

Direct observation of the superconducting gap in phonon spectra

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We show that the superconducting energy gap Δ can be directly observed in phonon spectra, as predicted by recent theories. In addition, since each phonon probes the gap on only a small part of the Fermi surface, the gap anisotropy can be studied in detail. Our neutron scattering investigation of the anisotropic conventional superconductor $\text{YNi}_2\text{B}_2\text{C}$ demonstrates this new application of phonon spectroscopy.

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In their seminal paper[1], Axe and Shirane found that the onset of superconductivity in Nb_3Sn affects the frequencies and the linewidths of phonons whose energies are comparable to the superconducting energy gap 2Δ . Similar effects were observed by neutron and Raman scattering in a number of conventional superconductors[2], but also in high- T_c copper oxides[3, 4, 5, 6, 7]. In many of these cases the authors argued that it was possible to determine whether the phonon energy was above or below 2Δ from the comparison of phonon frequencies and linewidths above and below T_c . Here we show that in the case of strong electron-phonon coupling, the entire phonon lineshape can change dramatically across T_c allowing a precise determination of 2Δ .

A few years ago, strong superconductivity-induced changes of the phonon lineshape were reported for a particular phonon in $\text{YNi}_2\text{B}_2\text{C}$ [8] and $\text{LuNi}_2\text{B}_2\text{C}$ [9], which - for the first time - could not be described by a simple change in the phonon linewidth. This observation stimulated further theoretical work[10, 11]. We performed very precise inelastic neutron scattering measurements of many different phonons in $\text{YNi}_2\text{B}_2\text{C}$ ($T_c = 15.2$ K) with a much smaller statistical error covering a much larger region of reciprocal space than in Ref. 8.

The neutron scattering experiments were performed on the 1T triple-axis spectrometer at the ORPHEE reactor at LLB, Saclay, and on the PUMA triple-axis spectrometer at the research reactor FRM II in Munich. Double focusing pyrolytic graphite monochromators and analysers were employed in both cases. A fixed analyser energy of 14.7 meV allowed us to use a graphite filter in the scattered beam to suppress higher orders. The experimental resolution was obtained from calculations using a formalism checked on phonons with very low coupling

strength. The wave vectors are given in reciprocal lattice units of $(2\pi/a \ 2\pi/b \ 2\pi/c)$, where $a = b = 3.51$ Å and $c = 10.53$ Å. The single crystal sample of $\text{YNi}_2\text{B}_2\text{C}$ weighing 2.26 g was grown by the floating zone method using the ^{11}B isotope to avoid strong neutron absorption. The single crystal sample was mounted in a standard orange cryostat at LLB and in a closed-cycle refrigerator at FRM II, allowing us measurements down to $T = 1.6$ K and 3 K, respectively.

Figure 1 shows the evolution through T_c of the low temperature lineshape of the phonon at the endpoint of the transverse acoustic branch in the (110) direction. This phonon has never been investigated before but its strong coupling to electrons has been predicted by density functional theory[12]. It appears at $\mathbf{q} = (0.5, 0.5, 0)$ (the M-point). On cooling from $T = 200$ K to a temperature just above $T_c = 15.2$ K, this phonon softens by about 10 % and broadens substantially, indicative of a strong electron-phonon coupling (data not shown). However, the lineshape remains Lorentzian to a very good approximation[18]. On further cooling through T_c , the lineshape starts to deviate strongly from a Lorentzian. In particular, a step-like increase at a certain energy E_s appears as the temperature is lowered through T_c (fig. 1, top). E_s increases with decreasing temperature below T_c . According to the theory developed by Allen et al. [10], a part of the low energy tail which lies below the gap, is pushed up in energy to form a narrow spike at 2Δ (fig. 1, bottom). The theory is based on the full quantum mechanical treatment of electron-phonon coupling where vibrational and electronic excitations mix into hybrid modes. The finite spectrometer resolution should wash out the spike in experiments resulting in a sharp intensity increase at 2Δ . The predicted intensity increase corresponds to the one we observe at E_s . We show below that other experimental features are also in an excellent agreement with this scenario. Thus we will interpret our results using the theoretical framework developed in Ref. 10.

A close comparison of the calculated lineshape with the

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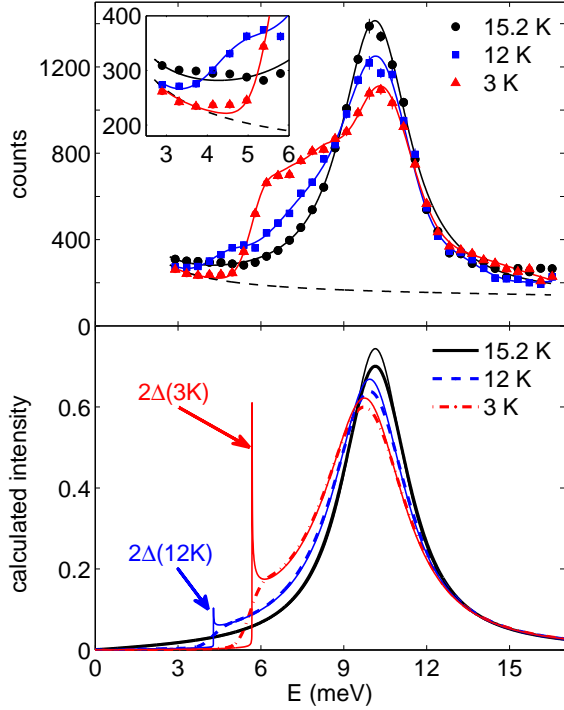


FIG. 1: (color online) *top*: Evolution of the neutron-scattering profile measured on $\text{YNi}_2\text{B}_2\text{C}$ at $\mathbf{Q} = (0.5, 0.5, 7)$ at $T = T_c$ (15.2 K) and below T_c . Solid lines are guides to the eye. The broken line denotes the background as determined from measurements at neighboring \mathbf{q} -points. *bottom*: Calculated phonon lineshapes based on the theory of Allen et al. [10], using parameters extracted from the experimentally observed lineshape in the normal state (thin lines). Thick lines are obtained after convolution with the experimental energy resolution.

low energy part of the observed phonon spectral function can give highly accurate values of 2Δ as a function of temperature (fig. 2, left). Close to T_c , where the step height is small, it is more convenient to scan the temperature at a fixed energy transfer: a sudden intensity decrease on cooling will mark the temperature where the gap is equal to the energy transfer (fig. 2, right).

The theory of Allen et al. [10] predicts not only the suppression of the spectral weight below 2Δ just mentioned, but also a downward shift of the intensity maximum and an intensity build-up on the low energy side of the peak (fig. 1, bottom). The observed changes are qualitatively similar, but are somewhat stronger than predicted. These discrepancies cannot be remedied by tuning the three parameters of the calculations. Two of them, the phonon energy and the phonon linewidth observed just above T_c , are entirely fixed by the phonon properties in the normal state and the third, the gap value is practically fixed as well by the sharp intensity increase at E_s . Notwithstanding the shortcomings of the actual theory - which might be due to the weak coupling

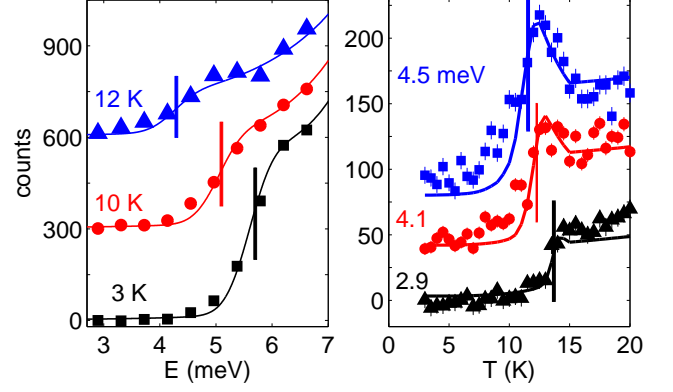


FIG. 2: (color online) *left*: Background subtracted energy scans taken at $\mathbf{Q} = (0.5, 0.5, 7)$ at three different temperatures including an offset of 300 counts for clarity. Lines were calculated from the theory of Allen et al. [10] after scaling to the experimentally observed step-heights and folding with the experimental energy resolution. The vertical bars indicate the gap values used in the calculations, respectively. *right*: Temperature scans taken at $\mathbf{Q} = (0.5, 0.5, 7)$ for three different energy transfers including an offset of 35 counts for clarity. Lines are as in the left panel. Vertical bars indicate the temperature for which the energy transfer is equal to the gap value 2Δ in the calculation.

approximation -, it predicts the observed changes with temperature over a wide range of energies remarkably well (fig. 2).

The features in the intensity vs. temperature curves (fig. 2, right) related to the gradual opening of the gap are quite sharp and, therefore, the gap values extracted from the data depend very little on fine details of the model. The temperature dependent gap value deduced in this way is plotted in fig. 4. The experimental curve deviates somewhat from the BCS weak coupling curve, which is not surprising for a superconductor with a T_c as high as 15.2 K. Likewise, the value of the zero temperature gap indicates strong coupling because the ratio $2\Delta/k_B T_c = 4.3$ (k_B is the Boltzmann constant) exceeds considerably the weak coupling limit of 3.52.

The method of extracting the superconducting gap from phonon lineshapes explained above looks straightforward. Therefore, one might ask why it has not yet been used so far. There are two stringent conditions for making the replica of the superconducting gap clearly visible: the phonon energy has to be considerably larger than 2Δ but on the other hand, the linewidth in the normal state has to be large enough that the tail of the phonon line extending into the gap region can still be discriminated from the background. One might also think to study phonons with an energy comparable to the gap energy. Here, the superconductivity-induced changes of the phonon lineshape are fairly strong, but extraction of the gap energy is possible only with the help of detailed calculations. An example for such a case is the acoustic phonon in the (100)-direction at wave vector $\mathbf{q} = (0.5,$

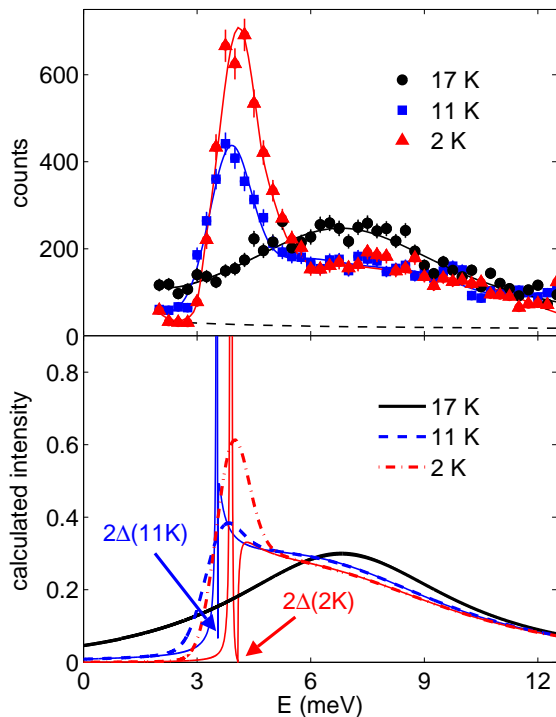


FIG. 3: (color online) *top*: Evolution of the neutron-scattering profile measured on $\text{YNi}_2\text{B}_2\text{C}$ at $\mathbf{Q} = (0.5, 0, 8)$ above and below T_c . Solid lines are guides to the eye. The broken line denotes the background as determined from measurements at neighboring \mathbf{q} -points. *bottom*: Calculated phonon lineshapes based on the theory of Allen et al.[10], using parameters extracted from the lineshape observed in the normal state (thin lines). The thick lines are obtained after convolution with the experimental resolution.

$0, 0)$. Profound changes of the lineshape of this phonon were first reported by Kawano et al.[8]. Puzzled by the appearance of an intense and relatively narrow line below T_c Kawano et al. thought to have found a new sharp excitation. We reinvestigated this phonon and show that the peculiar lineshapes appearing below T_c can be quite naturally understood using again the theory of Allen et al.[10].

The phonon at $\mathbf{q} = (0.5, 0, 0)$ has a significantly lower energy than the M-point phonon discussed above (7 meV as compared to 10 meV) and moreover, has a much larger linewidth[19]. As a consequence, the superconductivity-induced redistribution of spectral weight is even stronger, but the superconducting gap cannot be inferred from the data as easily, except for temperatures close to T_c where the gap is still small. For lower temperatures, most of the spectral weight condenses into a fairly sharp peak whose energy is somewhat below the gap. According to Ref. 10, this resonance can be regarded as a mixed vibrational/superelectronic collective excitation. Its position with respect to 2Δ has to be determined from calculations for the parameter values of this particular phonon. Again, the theory does not reproduce the observed line-

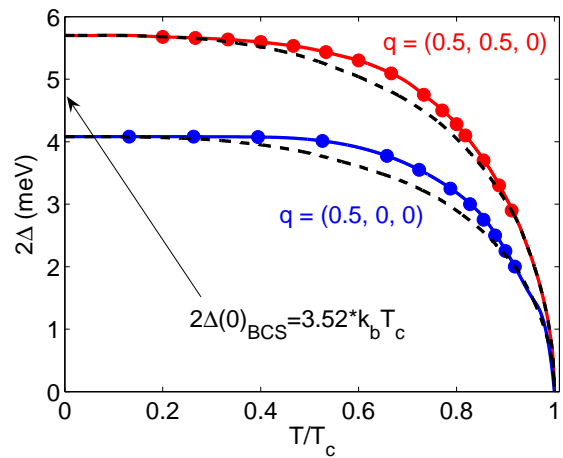


FIG. 4: (color online) Superconducting energy gap 2Δ versus temperature as evaluated from phonon data. Broken lines are calculated from weak-coupling BCS theory scaled to the experimental values at $T = 3$ K. Red and blue points were deduced from phonon measurements at different momentum transfers. Lines are guides to the eye.

shapes quantitatively, but sufficiently well to precisely determine the gap value (fig. 3).

Previous publications[1, 2] observed line narrowing for phonons with an energy smaller than 2Δ . We found that this temperature dependence can be quantitatively accounted for by the theory of Allen et al.[10]. Unfortunately, there are no phonons in $\text{YNi}_2\text{B}_2\text{C}$ with energies $< 2\Delta$, which have a sufficiently strong coupling to search for temperature effects of the linewidth in this compound.

The low temperature lineshapes of the two phonons discussed so far look quite distinct. A study of several phonons along the (100)-direction reveals that the lineshape may take on any intermediate form, depending on the phonon energy relative to the gap and on the normal state phonon linewidth. When going from $\mathbf{q} = (0.5, 0, 0)$ towards the zone boundary, the normal state phonon energy increases and the linewidth shrinks. As a result, the low temperature lineshape gradually evolves from that observed at $\mathbf{q} = (0.5, 0, 0)$ to that of the M-point phonon (fig. 5, (a)-(c)). Fig. 5 is interesting also for another reason: the superconductivity-induced redistribution of spectral weight involves the optic line at 14 meV as well with the spectral weight near both peaks transferred to much lower energies near the gap. At these \mathbf{q} -points, the acoustic and the optic modes strongly hybridize. Understanding this phenomenon in detail would require a highly nontrivial extension of the theory, which has not yet been made.

When plotting 2Δ extracted from the various phonons studied by us, it becomes clear that they probe different energy gaps, ranging from 4 meV to more than 6 meV (figs. 4, 5(d)). Phonons probe the gap on the parts of the Fermi surface connected by the phonon wave vector.

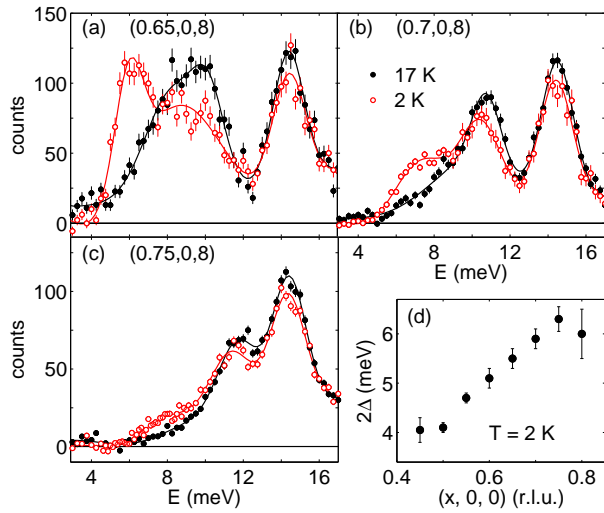


FIG. 5: (color online) Panels (a)-(c) show background subtracted energy scans taken at $\mathbf{Q} = (0.65-0.75, 0.8)$ at $T = 2$ K (open symbols) and 17 K (filled symbols). Lines are guides to the eye. Panel (d) describes the evolution of the low temperature superconducting gap value extracted from phonon scans along the (100) direction.

Only theory can tell which parts are actually involved. Density functional theory calculations may provide such information, but they require the technically involved solution of the anisotropic gap equations including the full momentum dependence of the electron-phonon coupling, which has not yet been carried out for $\text{YNi}_2\text{B}_2\text{C}$. In special cases, however, the respective parts of the Fermi surface can be inferred from the Fermi surface topology: for instance, the extremely large linewidth of the $\mathbf{q} = (0.5, 0, 0)$ phonon can be attributed to a nesting phenomenon predicted by theory[12].

Whereas the intensity step observed at $\mathbf{q} = (0.5, 0.5, 0)$ is very sharp, i.e. resolution-limited (≈ 1 meV), it is significantly broader (≈ 2 meV) at $\mathbf{q} = (0.7, 0, 0)$ and $(0.75, 0, 0)$, and therefore cannot be linked to a single, well-defined gap value. In this case the phonon wave vector may connect several different pieces of the Fermi surface probing a superposition of gap values.

Our results for the gap anisotropy in $\text{YNi}_2\text{B}_2\text{C}$ fit nicely to those extracted from tunnelling data using point-contact spectroscopy[13]. Further, the lowest gap seen in our phonon experiments agrees very well with the value derived from critical field $H_{c2}(T)$ data[14]. On the other hand, we have no evidence that the gap goes below $2\Delta = 4$ meV anywhere on the Fermi surface and possibly even to zero, as was proposed on the basis of field-angle-dependent heat capacity measurements[15] and field-angle dependent thermal conductivity data[16]. Extensive energy and wave vector resolved density functional theory calculations[12], which are in very good agreement with experiment[17], did not show any other suitable phonons in order to further probe the possible presence of nodes.

To conclude, we confirm the prediction of Allen et al. that the phonon line may acquire very peculiar shapes with prominent features directly related to the superconducting energy gap 2Δ . In contrast to a theoretical proposition[11], this effect is not restricted to phonons with wave vectors close to an extremum vector of the Fermi surface but may appear throughout the Brillouin zone. The gap value 2Δ can be directly inferred from the lineshape, often without recourse to a fit. This allows extraction of the gap values as a function of temperature with high accuracy. By studying phonons of different wave vectors, even the gap anisotropy can be explored by phonon spectroscopy. Thus phonon spectroscopy can be a much richer source of information on the superconducting properties than hitherto assumed.

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- D. Reznik, and K. Hradil, to be published soon.
- [18] The theoretical lineshape is not a true Lorentzian, but is very close to a Lorentzian for the case of the M-point phonon.
- [19] The lineshape of this phonon just above T_c is less well described by a Lorentzian because its linewidth is closer to the phonon energy.